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Simulation Modeling of Atmospheric Deposition of Sulfur from Biogenic and Anthropogenic Emissions

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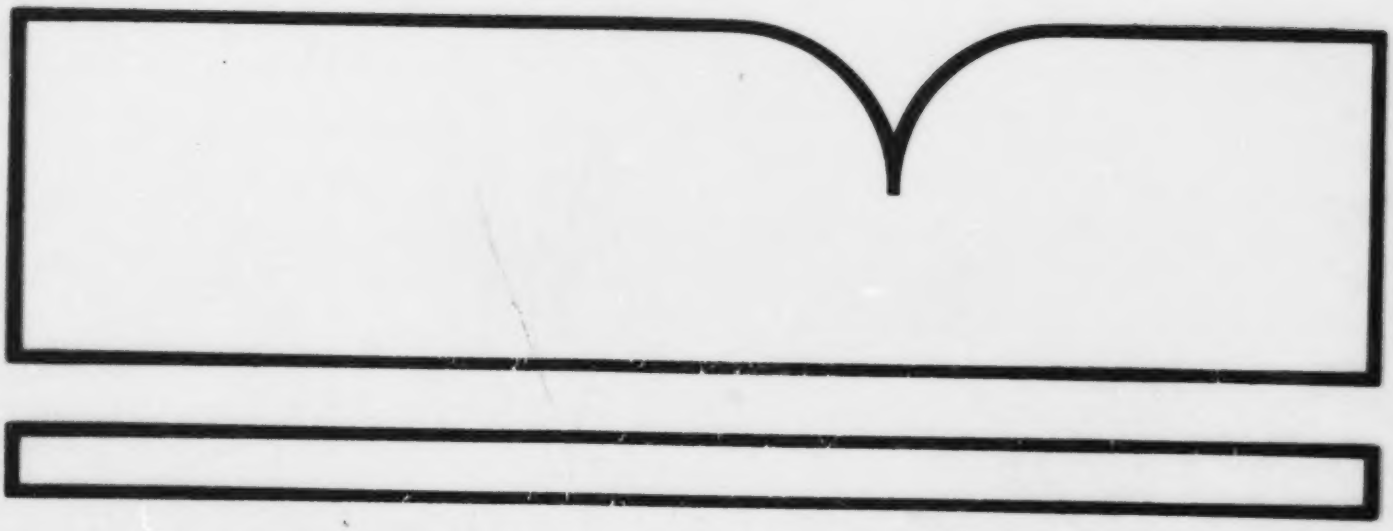

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SIMULATION MODELING OF ATMOSPHERIC DEPOSITION OF SULFUR FROM BIOGENIC AND ANTHROPOGENIC EMISSIONS

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A biogenic sulfur emission inventory for the eastern United States and immediately adjacent portions of Canada is used in the ASTRAP model to estimate biogenic contributions to wet and dry sulfur deposition and atmospheric sulfate concentration, under the conservative assumption that all biogenic sulfur is transformed to sulfate. Simulations for summer conditions, in which biogenic sulfur emissions are greatest, are compared with simulations of transport and deposition of anthropogenic emissions of sulfur. The biogenic contribution to sulfur deposition during summer is shown to have a relative maximum in the southeastern U.S., about 5 to 10% of the total deposition from combined anthropogenic and biogenic sources, with only a 1 to 2% biogenic contribution indicated in the northeastern U.S.

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SIMULATION MODELING OF ATMOSPHERIC DEPOSITION OF SULFUR FROM BIOGENIC AND ANTHROPOGENIC EMISSIONS

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A biogenic sulfur emission inventory for the eastern United States and immediately adjacent portions of Canada is used in the ASTRAP model to estimate biogenic contributions to wet and dry sulfur deposition and atmospheric sulfate concentration, under the conservative assumption that all biogenic sulfur is transformed to sulfate. Simulations for summer conditions, in which biogenic sulfur emissions are greatest, are compared with simulations of transport and deposition of anthropogenic emissions of sulfur. The biogenic contribution to sulfur deposition during summer is shown to have a relative maximum in the southeastern U.S., about 5 to 10% of the total deposition from combined anthropogenic and biogenic sources, with only a 1 to 2% biogenic contribution indicated in the northeastern U.S.

Simulations of long-range transport and deposition of acidifying pollutants have generally been confined to emissions from anthropogenic sources[1]. Biogenic and other natural emissions have been considered not amenable to control, less well quantified than anthropogenic emissions and, in the case of sulfur compounds, much smaller in magnitude over the northeastern United States than anthropogenic sources of sulfur.

State-of-the-art source-receptor models generally indicate that no single anthropogenic point source, however, contributes more than a few percent of the total deposition of anthropogenic sulfur at remote sites. Also, neither anthropogenic nor biogenic sources of atmospheric sulfur compounds are uniformly distributed[1,2]. Thus, the local contribution to sulfur deposition from biogenic sources may vary considerably from the contribution integrated over eastern North America as a whole, and may become large relative to the contributions from individual major point sources. (Here, "local" implies a scale of about 100 km, the approximate resolution of most simulation models of long-range transport and deposition).

In this study we compare the wet and dry sulfur deposition fields over the eastern United States resulting from biogenic emissions with the corresponding fields resulting from anthropogenic emissions. In addition, we compare the atmospheric sulfate concentration field associated with anthropogenic sources with an upper estimate of atmospheric sulfate associated with biogenic sources.

The simulation model of transport and deposition used here is the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model[1,3]. Summer conditions are simulated, since then the contribution of biogenic sources to sulfur deposition would be expected to be greatest, relative to other seasons, for both biological and meteorological reasons. Biogenic emissions exhibit a strong temperature dependence and are generally higher in the south, both because of higher temperatures and greater extent of wetlands; in addition, the prevailing wind patterns in the Southeast during summer exhibit a significant easterly to southeasterly component below the subtropical ridge of high pressure, and thus anthropogenic emissions from the industrial heartland are advected southward less frequently than during the cooler portions of the year.

In the trajectory module of the ASTRAP model, for either a point source or a virtual source representing an area, simulated tracers are released at 6-hr intervals in a time series of analyzed wind and precipitation fields for a period of one to three months. Each tracer initially represents a unit mass of pollutant emissions, which can be reduced by a function of the half power of any precipitation encountered. The ensembles of trajectories and wet deposition occurrences are described

statistically by mean positions and spreads of endpoints at plume-age (time since release) intervals of six hours out to seven days. There is no simulated short-term diffusion about any of the individual trajectories; rather, long-term diffusion results from the distributon of the trajectories about their mean path. (This is the origin of the "statistical" in the ASTRAP name.) A set of the statistics described above is calculated for each source or virtual source simulated.

A second module of ASTRAP numerically integrates a one-dimensional vertical profile. Dry deposition, vertical diffusion, and chemical transformation are treated in this second part. A unit emission is released in each specified layer (vertical resolution 100 m) in turn and diffused for seven days, with dry deposition velocities, linear first-order transformation rates, and profiles of vertical eddy diffusivity all varying diurnally and seasonally (but not spatially). Statistics are computed on surface concentration, airborne mass, and dry deposition mass increment for each emission layer at the same six-hour intervals for which trajectory statistics are stored.

A third ASTRAP module combines the statistics produced by the first two modules with an emission inventory and calculates the resulting concentration and deposition fields. By identifying grid elements as to underlying geopolitical entity, the deposition field can be integrated to produce deposition totals by state. The parameterization rates in ASTRAP are input options; averages of the key rates from summer simulations in this study are listed in Table I.

The assumption that biogenic emissions are instantly oxidized to sulfate is made here in order to provide an upper limit to the potential regional atmospheric sulfate burden from biogenic emissions. For consistency, the wet and dry removal rates used here for biogenic emissions are those associated with particulate sulfate: the proper removal rates for biogenic species are not known.

The estimates of biogenic sulfur emissions[2] are gridded with 80 km resolution over the original Sulfur Regional Experiment (SURE) region (roughly eastward and northward from Arkansas over the United States and adjacent portions of Canada). The report[2] also suggests appropriate values for a southward extension of the grid. In ASTRAP simulations the biogenic emissions are combined in 160 km grid cells.

TABLE I: ASTRAP PARAMETERIZATIONS

	Anthropogenic Emissions		Biogenic Emissions
	SO_2	SO_4	S
dry deposition velocity (average in cm/s)	0.45	0.25	0.25
wet removal (P = precip in cm/12 hr)	$\min P^{1/2}, 2/3$		same
transformation to SO_4 (average in % hr^{-1})	2.0	N/A	instantaneous
transport layer depth (m)	1500		same
emission horizontal resolution (km)	100-120		160
emission vertical resolution (m)	6 layers to 800 m		0-100

While anthropogenic emissions are thought to be better quantified than biogenic emissions, there is no universally accepted estimate of emissions in a form suitably disaggregated for model use. The inventories assembled as part of SURE[4] and the Multistate Atmospheric Power Production Pollution Study (MAP3S)[5] have desirable disaggregation, but the state totals, particularly in the SURE inventory, are generally higher than in the inventory of the Memorandum of Intent (MOI) on Transboundary Air Pollution studies[1]. The approach here is to use the disaggregated MAP3S inventory, but to scale the sources within each state in such a manner that the statewide total matches the MOI estimate. Uncertainty in model simulations attributable to emission uncertainties alone is likely about 10-20% in the results for anthropogenic emission discussed below.

Figures 1a through 1d are isopleths of the cumulative deposition fields for June through August of 1981 and the (potential) average atmospheric sulfate field resulting from biogenic emissions, while Figures 2a through 2d are the corresponding fields resulting from anthropogenic emissions. The biogenic fields have a maximum in the Southeast while the anthropogenic fields have a maximum in the upper Ohio Valley. Table 2 compares the integrated deposition totals for some states of particular interest. The biogenic share of sulfur deposition during summer is as high as 5% in Florida or 10% in Louisiana, but

only about 1% in the northeastern state of New York. The total biogenic emissions during summer are about 1.5% of the total anthropogenic emissions simulated, but the biogenic emission inventory is for a smaller area, essentially that plotted as a map background. Since the ratio of biogenic emissions to anthropogenic emissions is about twice as high during summer as it is for the year as a whole, a rough estimate of the biogenic relative contribution to annual sulfur deposition would be about one half of that in summer simulations.

TABLE II. SULFUR DEPOSITION (WET PLUS DRY) FOR SELECTED STATES FOR JUNE THROUGH AUGUST, 1981 METEOROLOGICAL CONDITIONS

	Total Sulfur Deposition (kT/Season)	
	Biogenic Sources	Anthropogenic Sources
Florida	1.3	25
Louisiana	0.7	7
Mississippi	0.8	16
Alabama	1.0	34
Georgia	1.6	50
South Carolina	0.7	24
Tennessee	0.8	54
New York	0.7	78

Biogenic Emissions Sure Area and Southward	49 kT/Season
Anthropogenic Emissions U.S.A. and Canada	3500 kT/Season

The anthropogenic emissions inventory used here covers all of the U.S. and Canada: the biogenic emissions inventory covers only the states contiguous to or eastward of the Mississippi River, plus adjacent parts of Canada and coastal waters. Exclusion of biogenic emissions from other parts of the continent results in an underestimate of the biogenic contribution to the western and northern portions of the calculation grid: the effect is probably greatest in absolute terms in Louisiana, because of the higher emissions likely in nearby east Texas wetlands.

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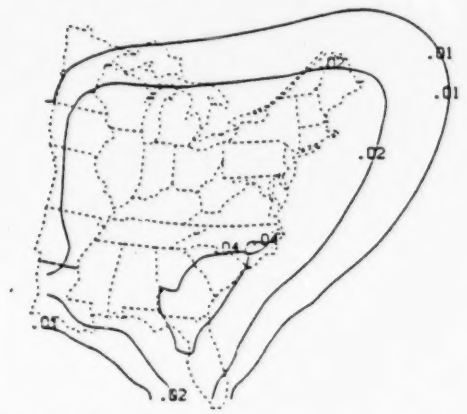
Figure 1. Isopleths of (a) sulfur wet deposition, (b) sulfur dry deposition, (c) wet plus dry deposition, and (d) atmospheric sulfate concentration for June through August 1981 associated with biogenic emissions.

Figure 2. Isopleths of (a) sulfur wet deposition, (b) sulfur dry deposition, (c) wet plus dry deposition, and (d) atmospheric sulfate concentration for June through August 1981 associated with anthropogenic emissions.

WET DEPOSITION OF BIOGENIC SULFUR

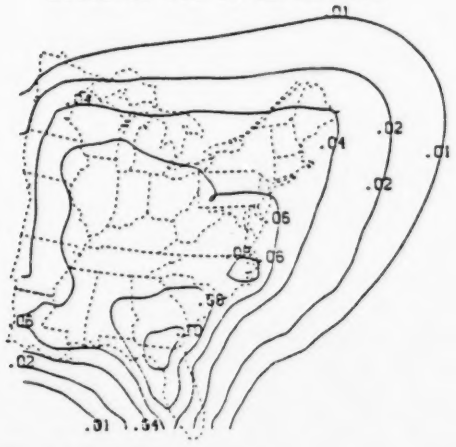


BIOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 0.05 KG SULFUR PER HECTARE

DRY DEPOSITION OF BIOGENIC SULFUR

BIOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 0.10 KG SULFUR PER HECTARE

BIOGENIC SULFUR DEPOSITION

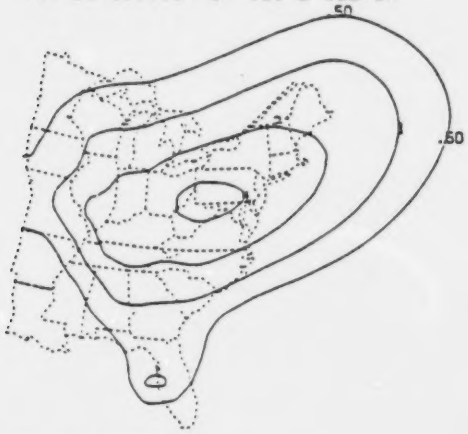


BIOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 0.14 KG SULFUR PER HECTARE

ATMOSPHERIC SULFATE

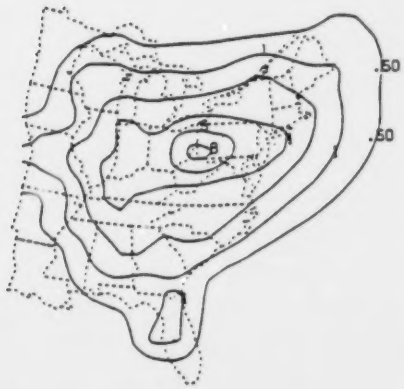
BIOGENIC EMISSIONS
SUMMER 1981 AVERAGE CONCENTRATION
MAX - 1.41 µG PER CUBIC METER

WET DEPOSITION OF TOTAL SULFUR



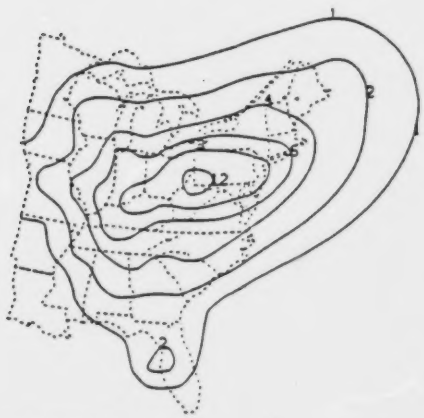
ANTHROPOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 5.95 KG SULFUR PER HECTARE

DRY DEPOSITION OF TOTAL SULFUR



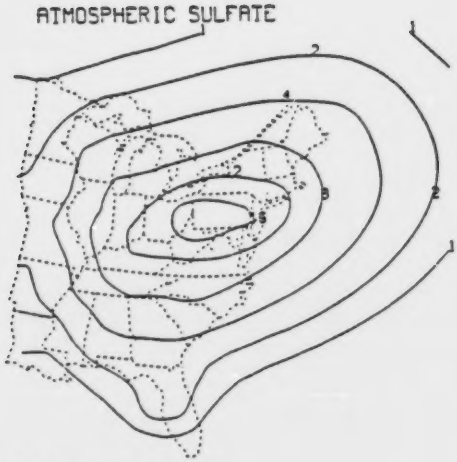
ANTHROPOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 10.9 KG SULFUR PER HECTARE

TOTAL SULFUR DEPOSITION



ANTHROPOGENIC EMISSIONS
SUMMER 1981 ACCUMULATION
MAX - 16.6 KG SULFUR PER HECTARE

ATMOSPHERIC SULFATE



ANTHROPOGENIC EMISSIONS
SUMMER 1981 AVERAGE CONCENTRATION
MAX - 20.8 µG PER CUBIC METER